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Development of procedures, devices, and experimental setup for a microfluidic investigation of Underground Hydrogen Storage

Abstract

Recognizing the intricate link between macro-scale phenomena and behaviors at smaller scales, this research aimed to develop a microfluidic approach for the investigation of fluid flow phenomena at the micro-scale, occurring within porous media representative of geological formations adopted for Underground Fluid Storage. Microfluidic devices, which are transparent and engraved to replicate rock microstructure, allow for direct observation of fluid dynamics, offering a significant advantage over traditional core analysis methods. These devices are reusable, replicable, and allow for precise adjustments of geometric parameters and surface properties. They also require minimal material and fluid use and are relatively simple to manufacture. The microfluidic patterned core simulates the porous network, and fluid dynamics are directly observable and recordable using an optical microscope and a camera. Inlet and outlet channels connect to external microfluidic pumps and tubing for fluid injection and extraction.

This PhD work represented an effort to build a microfluidic experimental approach for the investigation of multiphase flow in porous systems in the context of underground gas storage. More specifically, the goal was to develop a method to investigate pore-scale dynamics occurring during the injection and withdrawal operations of hydrogen in depleted gas reservoirs. If the micro-scale phenomenology of natural gas underground systems is not yet fully understood, much less is known about non-native fluids' behavior and phase interactions. Therefore, microfluidic studies applied to underground hydrogen storage systems may help fill up a present research gap.

Following the tasks defined in collaboration with SNAM-Stogit, we set a broad spectrum of laboratory activities which laid out a promising research path. We successfully achieved to:

1) Design a set of 17 microfluidic patterns modelling porous media, having inlet channels that guarantee a uniform fluid front at the entrance of the porous pattern. The designed patterns followed incremental complexity: two regular patterns– two Voronoi diagrams– three QSGS-generated media– ten mosaics of real rock binarized images.
2) Optimize the fabrication process of soft-lithography for PMDS-glass devices, and of Deep Reactive Ion Etching for Silicon-glass chips, and fabricate 12 PDMS-glass devices, hosting 4 different patterns and 8 silicon-glass devices hosting a regular pattern.
3) Design and implement the experimental set-up for the fluid flow tests (Fig.1). We purchased the components of the experimental apparatus considering the necessities of the designed future work. Commercial devices by Micronit® were also purchased. In particular, the set-up was designed for hydrogen handling and high-pressure tests.
4) Validate the fabrication processes and the experimental apparatus by preliminary fluid flow tests (Fig.2), aimed at: i) observing pore-scale phenomena and percolation patterns inside diverse porous structures; ii) extracting the absolute permeability of the porous system

through monophase fluidic tests; iii) extracting the relative permeability of the phases for gas drainage and water imbibition.

The preliminary multiphase tests for pore-scale observations (i) clearly showed the dependency of the percolation forms on the system, given the same capillary number and viscosity ratio. This introduces an interesting discussion around the role of the capillary number in comparing different systems. As described by the Lenormand diagram, the percolation patterns depend on the structure of the porous medium. We could observe a difference between the percolation patterns at $Ca=10^{-5}$ and Ca 10^{-4}, with similarities across the patterns for the same Ca; however, it was not trivial to compare very different geometries. The regular pattern and the Voronoi diagram, due to little heterogeneity in terms of pore-throat dimensions, grain size and tortuosity, show few pore-scale phenomena; the Hostun pattern allowed for the visualization of Haines jumps, and the viscous fingering was more evident. The QSGS, artificially elongated in an anisotropic structure, also showed viscous fingering and bypassing. In all cases, most of the displacement occurred in the channels aligned with the fluid flow. The residual saturations were always smaller for $Ca=10^{-4}$, but we could not make sure to have entered a constant-saturation domain comparing two Ca only. More Ca should be explored to draw the constant-saturation domains and the transition zones for the systems. This would help understand how the velocity domains affect the residual saturations, which in turn are a measure of the storage capacity of the system. The absolute permeability measurement (ii) reported optimal results in terms of reproducibility and agreed with the measurements provided by literature works. The relative permeability measurement (iii) helped us validate the set-up designed to inject.

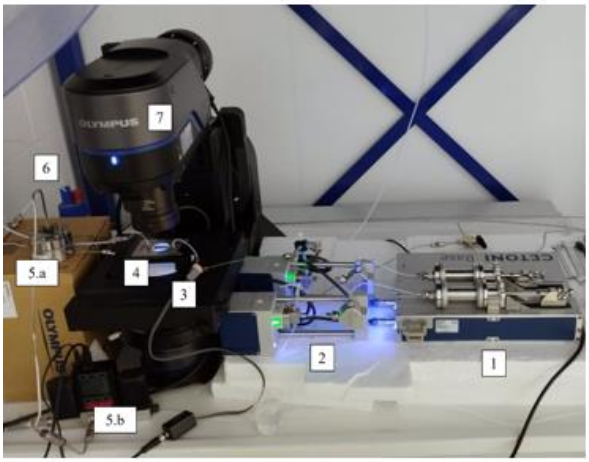

Figure 1. A picture of the setup. 1) CETONI Nemesys M Syringe pumps; 2) Three-way Cetoni Contiflow valves; 3) Elveflow MPS; 4) Micronit device and High-pressure reactor; 5.a) Back pressure regulator; 5.b) Electronic BPR controller; 6) Collecting bottles; 7) Olympus DSX1000. The water reservoir is next to the three-way valves, the gas tank is outside the view of the picture.

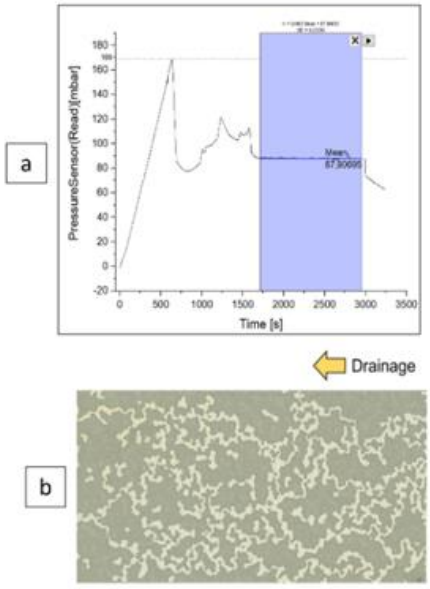

Figure 2. a) Example of pressure measurement during the drainage tests performed with hydrogen as the non-wetting fluid. b) Map of the device at the end of the drainage test, when the pressure has stabilized but the fluids are still flowing, i.e., the flow conditions are dynamic